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Josephson Effect due to Odd-frequency Pairs in Diffusive Half Metals

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The Josephson effect in superconductor / diffusive ferromagnet / superconductor (SFS) junctions is studied using the recursive Green function method in the regime of large exchange energy in a ferromagnet. Motivated by recent experiment [R. S. Keizer, *et. al.*, Nature **439**, 825 (2006)] we also address the case of superconductor / diffusive half metal / superconductor junctions. The pairing function in spin-singlet and triplet channels, the Josephson current and their mesoscopic fluctuations are calculated. We show that the spin-flip scattering at the junction interfaces opens the Josephson channel of the odd-frequency spin-triplet Cooper pairs. As a consequence, the local density of states in half metals has a large peak at the Fermi energy. Therefore odd-frequency pairs can be detected experimentally by using the scanning tunneling spectroscopy.

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Ferromagnetism and spin-singlet superconductivity are competing orders against each other because the exchange field breaks down the spin-singlet pairs. The Cooper pairs, however, do not always disappear under exchange fields. The Fulde-Ferrell-Larkin-Ovchinnikov[1,2] (FFLO) state and the proximity effect in ferromagnets[3,4,5,6,7,8,9,10] are typical examples. Under exchange fields, the pairing function oscillates and changes its sign in the real space. As a consequence, superconductor / ferromagnet / superconductor (SFS) junctions undergo the 0-π transition with varying length of a ferromagnet or temperature. It was also predicted that the odd-frequency spin-triplet pairs appear in weakly polarized ferromagnets with rotating magnetization direction near the junction interface[6]. These pairs have long range penetrarion into ferromagnets[6,7]. Thus Cooper pairs change their original face to survive under exchange fields.

Half metal is an extreme case of completely spin polarized material because its electronic structure is insulating for one spin direction and metallic for the other. At a simple thought, the spin-singlet Cooper pairs would not be able to penetrate into half metals. However recent experiment[11] showed the existence of the Josephson coupling in superconductor / half metal / superconductor (S/HM/S) junctions, where NbTiN was used as a s-wave superconducting electrode and CrO_2 as a half metal. Thus one has to seek a new state of Cooper pairs in half metals attached to spin-singlet superconductors. Prior to the experiment[11], Eschrig *et. al.*[12] have addressed this challenging issue. In the *clean limit*, they showed that the p-wave spin-triplet pairs induced by the spin-flip scattering at the interface can carry the Josephson current. In real S/HM/S junctions, however, half metals are in the diffusive transport regime; the elastic mean free path of an electron is much smaller than the size of a half metal. In addition, real S/HM/S junctions are close to the dirty limit because the coherence length in a ferromagnet may become comparable to the mean free path. In such diffusive half metals, the p-wave symmetry of Cooper pairs is not possible because the pair wave function is isotropic in the momentum space due to impurity scattering[13]. In this paper, we provide a general theory for the Josephson effect in diffusive SFS junctions with arbitrary magnitude of the exchange field V_{ex}. When V_{ex} is much larger than the superconducting pair potential at zero temperature Δ_0, mesoscopic fluctuations of the Josephson current are much larger than the ensemble averaged value. In addition, we focus on interesting case of diffusive S/HM/S junctions. We show that the odd-frequency spin-triplet s-wave pairing state is realized in half metals and propose an experimental method to detect this property.

Let us consider the two-dimensional tight-binding model for a SFS junction as shown in Fig. 1(a). The vector $\boldsymbol{r} = j\boldsymbol{x} + m\boldsymbol{y}$ points to a lattice site, where $\boldsymbol{x}$ and $\boldsymbol{y}$ are unit vectors in the x and y directions, respectively. In the y direction, we apply the periodic boundary condition for the number of lattice sites being W. Electronic states in superconducting junctions are described by the mean-field Hamiltonian

$$\begin{aligned} H_{\rm BCS} =& \frac{1}{2}\sum_{\boldsymbol{r},\boldsymbol{r}'}\left[\tilde{c}^\dagger_{\boldsymbol{r}}\;\hat{h}_{\boldsymbol{r},\boldsymbol{r}'}\;\tilde{c}_{\boldsymbol{r}'} - \overline{\tilde{c}_{\boldsymbol{r}}}\;\hat{h}^*_{\boldsymbol{r},\boldsymbol{r}'}\;\overline{\tilde{c}^\dagger_{\boldsymbol{r}'}}\right] \\ &+ \frac{1}{2}\sum_{\boldsymbol{r}\in \mathrm{S}}\left[\tilde{c}^\dagger_{\boldsymbol{r}}\;\hat{\Delta}\;\overline{\tilde{c}^\dagger_{\boldsymbol{r}}} - \overline{\tilde{c}_{\boldsymbol{r}}}\;\hat{\Delta}^*\;\tilde{c}_{\boldsymbol{r}}\right], \end{aligned} \tag{1}$$

$$\begin{aligned} \hat{h}_{\boldsymbol{r},\boldsymbol{r}'} =& \left[-t\delta_{|\boldsymbol{r}-\boldsymbol{r}'|,1} + (\epsilon_{\boldsymbol{r}} - \mu + 4t)\delta_{\boldsymbol{r},\boldsymbol{r}'}\right]\hat{\sigma}_0 \\ &-\boldsymbol{V}(\boldsymbol{r})\cdot\hat{\boldsymbol{\sigma}}\delta_{\boldsymbol{r},\boldsymbol{r}'}, \end{aligned} \tag{2}$$

with $\overline{\tilde{c}}_{\boldsymbol{r}} = (c_{\boldsymbol{r},\uparrow}, c_{\boldsymbol{r},\downarrow})$, where $c^\dagger_{\boldsymbol{r},\sigma}$ ($c_{\boldsymbol{r},\sigma}$) is the creation (annihilation) operator of an electron at $\boldsymbol{r}$ with spin $\sigma = (\uparrow$ or $\downarrow)$, $\overline{\tilde{c}}$ means the transpose of $\tilde{c}$, $\hat{\sigma}_l$ for $l = 1-3$ are the Pauli's matrices, and $\hat{\sigma}_0$ is 2×2 unit matrix. The hopping integral t is considered among nearest neighbor sites in both superconductors and ferromagnets. In a ferromagnet, the on-site scattering potentials are given randomly in the range of $-V_I/2 \leq \epsilon_{\boldsymbol{r}} \leq V_I/2$ and the uniform exchange potential is given by $\boldsymbol{V}(\boldsymbol{r}) = V_{ex}\boldsymbol{e}_3$, where $\boldsymbol{e}_l$ for $l = 1-3$ is unit vector in a spin space. The Fermi energy μ is set to be $2t$ in a normal metal with $V_{ex} = 0$, while a ferromagnet and a half metal are respectively described by V_{ex}/t = 1 and 2.5 in Fig. 1(b). The spin-flip scatterings are introduced at $j = 1, 2$, $L_N - 1$, and

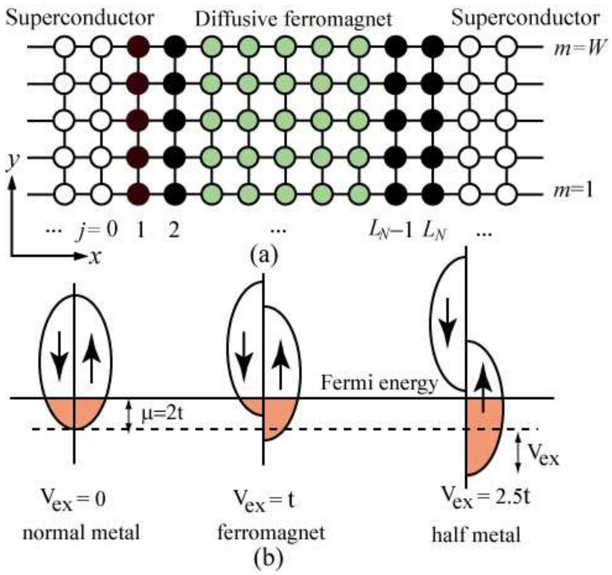

FIG. 1: (Color online) (a): A schematic figure of a SFS junction on the tight-binding lattice. (b): The density of states for each spin direction. The Josephson junction is of the SNS, SFS, and S/HM/S type for $V_{ex}/t = 0$, 1 and 2.5, respectively.

L_N, where we choose $\boldsymbol{V}(\boldsymbol{r}) = V_S \boldsymbol{e}_2$. In superconductors we take $\epsilon_{\boldsymbol{r}} = 0$ and choose $\hat{\Delta} = i\Delta\hat{\sigma}_2$, where Δ is the amplitude of the pair potential in the s-wave symmetry channel.

The Hamiltonian is diagonalized by the Bogoliubov transformation and the Bogoliubov-de Gennes (BdG) equation is numerically solved by the recursive Green function method[14,15]. We calculate the Matsubara Green function,

$$\check{G}_{\omega_n}(\boldsymbol{r},\boldsymbol{r}') = \begin{pmatrix} \hat{g}_{\omega_n}(\boldsymbol{r},\boldsymbol{r}') & \hat{f}_{\omega_n}(\boldsymbol{r},\boldsymbol{r}') \\ -\hat{f}^*_{\omega_n}(\boldsymbol{r},\boldsymbol{r}') & -\hat{g}^*_{\omega_n}(\boldsymbol{r},\boldsymbol{r}') \end{pmatrix}, \tag{3}$$

where $\omega_n = (2n+1)\pi T$ is the Matsubara frequency, n is an integer number, and T is a temperature. The Josephson current is given by

$$J = -ietT \sum_{\omega_n} \sum_{m=1}^{W} \mathrm{Tr}\left[\check{G}_{\omega_n}(\boldsymbol{r}',\boldsymbol{r}) - \check{G}_{\omega_n}(\boldsymbol{r},\boldsymbol{r}')\right] \tag{4}$$

with $\boldsymbol{r}' = \boldsymbol{r} + \boldsymbol{x}$. In this paper, 2×2 and 4×4 matrices are indicated by $\hat{\cdots}$ and $\check{\cdots}$, respectively. The quasiclassical Green function method is a powerful tool to study the proximity effect. However the quasiclassical Green function cannot be constructed in a half metal because the Fermi energy is no longer much larger than the pair potential for one spin direction. On the other hand, there is no such difficulty in our method. In addition, it is possible to obtain the ensemble average of the Josephson current $\langle J\rangle = (1/N_S)\sum_{i=1}^{N_S} J_i$ and its fluctuations $\delta J = \sqrt{\langle J^2\rangle - \langle J\rangle^2}$ after calculating Josephson current in a large number of samples N_S with different impurity configurations. These are the advantages of the recursive Green function method. Throughout this paper we fix the following parameters: $L_N = 74$, $W = 25$, $\mu = 2t$, $V_I = 2t$ and $\Delta_0 = 0.005t$ [16]. This parameter choice corresponds to the diffusive transport regime in the N, F and HM layers. The results presented below are not sensitive to variations of these parameters.

We first discuss the Josephson current in SFS junctions as shown in Fig. 2(a) for $T = 0.1T_c$ where T_c is the transition temperature. We assume that the spin-flip scattering at the interfaces is absent (i.e., $V_S = 0$) and fix the phase difference across the junctions φ equal to $\pi/2$. The results are normalized by $\langle J_0\rangle$ which is the Josephson current in superconductor / normal metal / superconductor (SNS) junctions (i.e., $V_{ex} = 0$). We define the coherence length $\xi_h = \sqrt{D/2V_{ex}}/a_0$ measured in units of lattice constant a_0 with D being the diffusion coefficient. The Josephson current oscillates as a function of V_{ex} and changes its sign almost periodically. The sign changes of $\langle J\rangle$ correspond to the 0-π transitions in an SFS junction. At the same time, the amplitude of $\langle J\rangle$ decreases rapidly with increasing V_{ex}. We should pay attention to the relation $\langle J\rangle \ll \delta J$ which means that the Josephson current is not the self-averaging quantity. It is impossible to predict the Josephson current in a single sample J_i from the ensemble average $\langle J\rangle$ because J_i strongly depends on a microscopic impurity configuration. In fact, the Josephson current flows in *a single sample* even if $\langle J\rangle = 0$ at the transition points. Roughly speaking, $\langle J\rangle$ vanishes because half of samples are 0-junctions and the rest are π-junctions[18]. Since $\langle J\rangle = 0$,

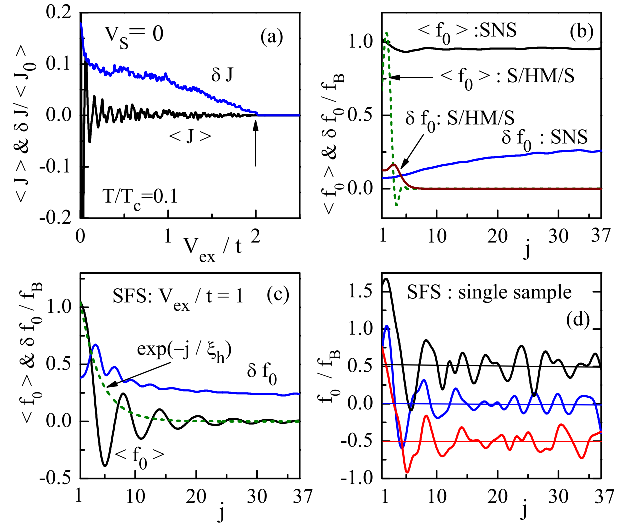

FIG. 2: (Color online) (a) The Josephson current versus the exchange potential V_{ex}. At $V_{ex} = 2t$ ferromagnets become half-metallic as indicated by an arrow. (b) The pairing function of spin-singlet pairs and its fluctuations versus position j for $V_{ex}/t = 0$ (SNS) and 2.5 (S/HM/S); (c) for $V_{ex}/t = 1$ (SFS). (d) The pairing function in three different samples for $V_{ex}/t = 1$, the vertical axis is offset by 0.5 as indicated by the horizontal lines. The spin-flip scattering is absent in all panels, $V_S = 0$.

δJ approximately corresponds to the typical amplitude of the Josephson current expected in a single sample. The relation $\langle J\rangle = 0$ has different meaning for SFS and S/HM/S cases. In SFS junctions, $\langle J\rangle = 0$ at the transition points is the result of the ensemble averaging and the Josephson current remains finite in a single sample. The characteristic temperature and length of a ferromagnet at the $0-\pi$ transitions vary from one sample to another. In S/HM/S junctions at $V_{ex} = 2.5t$, however, $\langle J\rangle = 0$ means vanishing Josephson current even in a single sample[12] because $\langle J\rangle = \delta J = 0$.

The origin of large fluctuations of the Josephson current can be understood by considering the behavior of the pairing function in a ferromagnet. The pairing function in Eq. (3) can be decomposed into four components,

$$\frac{1}{W}\sum_{m=1}^{W} \hat{f}_{\omega_n}(\boldsymbol{r},\boldsymbol{r}) = i\sum_{\nu=0}^{3} f_\nu(j)\hat{\sigma}_\nu\hat{\sigma}_2, \tag{5}$$

where $f_0(f_3)$ is the pairing function of the spin-singlet (spin-triplet) pairs with the spin structure of $(|\uparrow\downarrow\rangle - (+)\,|\downarrow\uparrow\rangle)/\sqrt{2}$, respectively, and the pairing function of $|\uparrow\uparrow\rangle$ ($|\downarrow\downarrow\rangle$) is given by $f_{\uparrow\uparrow} = if_2 - f_1$ ($f_{\downarrow\downarrow} = if_2 + f_1$). In Figs. 2(b) and (c), we show $\langle f_0\rangle$ and δf_0 as a function of position j in a diffusive ferromagnet, where f_B is the pairing function in bulk superconductor, ω_n is fixed at $0.02\Delta_0$, $V_S = 0$, and $\varphi = 0$. The junction interface and the center of a ferromagnet correspond to $j = 1$ and $j = 37$, respectively. In SNS junctions (Fig. 2b), $\langle f_0\rangle$ is larger than δf_0 and very weakly decays with j. The spin-singlet Cooper pairs exist everywhere in a normal metal. Near the interface, δf_0 is slightly suppressed due to the tight contact to the superconductor. On the other hand, in SFS junctions in (Fig. 2c), the average $\langle f_0\rangle$ decreases exponentially with j according to $\exp(-j/\xi_h)$ as indicated by a broken line. The fact that δf_0 remains finite at the center of a ferromagnet means that the spin-singlet pairs penetrate far beyond ξ_h even though $\langle f_0\rangle \sim 0$ there.

In Fig. 2(d), we show the pairing function for three different realization of disorder in SFS junctions. The pairing functions are in phase near the interface ($j \le \xi_h$), whereas they are out of phase far from the interface. We obtain the relation $\delta f_0 \propto e^{-j/\xi_T}$ with $\xi_T = \sqrt{D/2\omega_n}$ in agreement with Ref. 17. Thus we conclude that spin-singlet Cooper pairs do exist in *a single sample* of ferromagnet even for $j \gg \xi_h$ and the mesoscopic fluctuations of the pairing function provide the origin of the large fluctuations in the Josephson current. In S/HM/S junctions for $V_{ex} = 2.5t$ as shown in Fig. 2(b), both $\langle f_0\rangle$ and δf_0 vanish for $j \gg 1$, which indicates the absence of spin-singlet Cooper pairs in a half metal.

The relation $\langle J\rangle \ll \delta J$ is the characteristic feature of the Josephson current in diffusive SFS junctions. This feature, however, is drastically changed by the spin-flip scattering at the interfaces. In Figs. 3 (a) and (b) we show $\langle J\rangle$ and δJ *vs* V_S for $V_{ex}/t = 1$ and 2.5, respectively. In both cases (a) and (b), we find that $|\langle J\rangle| \ge \delta J$ for $V_S \ge 0.3t$. The Josephson current becomes self-averaging in the presence of the spin-flip scattering. The reason can be explained by calculating the pairing functions of equal-spin pairs shown in Figs. 3(c) and (d), where f_ν is plotted as a function of position j. Here we

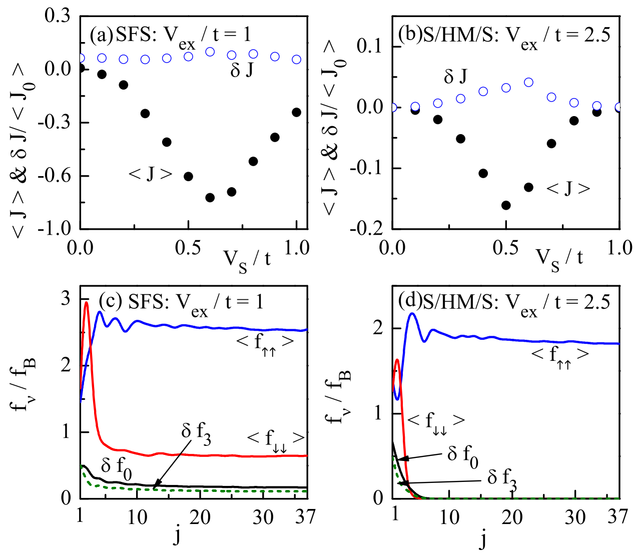

FIG. 3: (Color online) (a) The Josephson current and its fluctuations at $T = 0.1T_c$ and $\varphi = \pi/2$ as a function of the interface spin-flip scattering V_S for $V_{ex}/t = 1$ and (b) for $V_{ex}/t = 2.5$. The pairing functions versus position j in a ferromagnet (c) and in a half metal (d) at $V_s = 0.4t$, $\varphi = 0$ and $\omega_n = 0.02\Delta_0$. In (d), differences among $\langle f_{\downarrow\downarrow}\rangle$, δf_0 and δf_3 at large j become small.

show δf_0 and δf_3 instead of $\langle f_0\rangle$ and $\langle f_3\rangle$ because the ensemble averages are much smaller than their fluctuations. The fast decay of δf_0 and δf_3 is determined by strong spin polarization. In both cases (c) and (d), $\langle f_{\uparrow\uparrow}\rangle$ becomes larger than δf_0 and δf_3 because the pairing function $f_{\uparrow\uparrow}$ does not change sign for various impurity configurations. Thus the averaged quantities become larger than their fluctuations. Thus the Josephson current becomes self-averaging as shown in Figs. 3(a) and (b).

Finally we address an unusual symmetry property of the Josephson current in S/HM/S junctions. In Fig. 4 (a), we show $\langle f_{\uparrow\uparrow}\rangle$ as a function of ω_n, where $j = 37$, $V_S = 0.2t$, $\varphi = 0$, and $V_{ex} = 2.5t$. For comparison, we also show $\langle f_0\rangle$ on the normal side of a SNS junction. The pairing function $\langle f_0\rangle$ in a normal metal is the even function of ω_n, whereas $\langle f_{\uparrow\uparrow}\rangle$ in a half metal is the odd function of ω_n Ref. 6. The pairing function obeys the Pauli's rule

$$\hat{f}_{\omega_n}(\boldsymbol{r},\boldsymbol{r}') = -\bar{\hat{f}}_{-\omega_n}(\boldsymbol{r}',\boldsymbol{r}), \tag{6}$$

where $\bar{\hat{f}}$ denotes the transpose of $\hat{f}$ meaning the interchange of spins. It is well known that ordinary even-frequency pairs are classified into two symmetry classes: the spin-singlet even-parity and the spin-triplet odd-parity one. In the former case, the negative sign arises due to the interchange of spins, while in the latter case due to $\boldsymbol{r} \leftrightarrow \boldsymbol{r}'$. In the present calculation, all components on the right hand side of Eq. (5) have

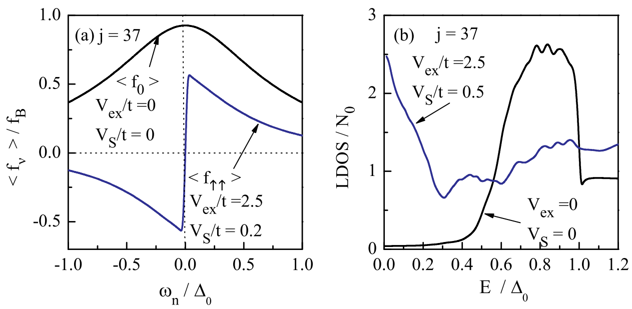

FIG. 4: (Color online) (a): Dependences of the pairing functions on ω_n. (b): The local density of states at $j = 37$ in a half metal at $V_S = 0.5t$ and in a normal metal at $V_S = 0$.

the s-wave symmetry. The pairing functions are isotropic in both the real and momentum spaces due to diffusive impurity scattering. As a result, $f_{\uparrow\uparrow}$ must be the odd function of ω_n to obey the Pauli's rule. Both even- and odd-frequency pairs are mixed in ferromagnets as shown in Fig. 3(c). The fraction of odd-frequency pairs depends on parameters such as the exchange potential and the spin-flip scattering. On the other hand, in a diffusive half metal all Cooper pairs have the odd-frequency character, which causes drastic change in the quasiparticle density of states.

The density of states is given by $N(E, j) = -\frac{1}{\pi}\frac{1}{W}\sum_{m=1}^{W} \mathrm{ImTr}\check{G}_{E+i\gamma}(\boldsymbol{r}, \boldsymbol{r})$, where γ is a small imaginary part chosen to be $0.05\Delta_0$ in the following. In Fig. 4(b), the local density of states (LDOS) at $j = 37$ is shown, where $\varphi = 0$ and N_0 is the density of states in the normal state at $V_{ex} = 0$. For comparison, we show LDOS on the normal side of SNS junction with $V_S = 0$ which has a minigap at $E < E_{Th} \sim 0.3\Delta_0$, where E_{Th} is the Thouless energy. In contrast to that, LDOS in a half metal has a peak at the Fermi energy, it's width is characterized by E_{Th}. This peak is generated at the spin active interface by the mechanism discussed in Ref 20 and is tranfered into a half metal due to long range property of odd-frequency spin-triplet even-parity pairing function. The peak is much stronger than the enhancement of the LDOS found in weak ferromagnets[3,10,21,22]. In addition, in a half metal the peak shape is almost independent of position, while in the SF junctions[21] the LDOS has an oscillatory peak/dip structure at $E = 0$ which rapidly decays with the distance from the SF interface. Therefore the large peak at $E = 0$ in LDOS is a robust and direct evidence of the odd-frequency pairing in half metals. To test the existence of such peculiar pairing state, the scanning tunneling spectroscopy could be used.

In conclusion, we have studied Josephson effect in superconductor / diffusive ferromagnet / superconductor junctions by using the recursive Green function method. The Josephson current in these junctions basically is not self-averaging because the spin-singlet Cooper pairs penetrating into ferromagnets far beyond ξ_h cause the large fluctuations of the pairing function. In the presence of the spin-flip scattering at the interfaces, the equal-spin odd-frequency pairs drastically suppress the fluctuations. When ferromagnets are half-metallic, all Cooper pairs have the odd-frequency property. As a result, the low energy peak in the quasiparticle density of states in a half metal exists at the distances far beyond ξ_h from the interface and could be probed by scanning tunneling spectroscopy.

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